

REPORT DOCUMENTATION PAGE

AFRL-SR-AR-TR-02-

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1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE	3. REPORT TYPE AND DATES COVERED 01 Dec 98 to 30 Nov 01 FINAL	
4. TITLE AND SUBTITLE New Materials and Structures for Nonlinear Optics			5. FUNDING NUMBERS 61102F 2301/AX	
6. AUTHOR(S) Professor Boyd				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) University of Rochester 518 Hylan Building Rochester NY 14627-0140			8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NE 801 North Randolph Street Rm 732 Arlington, VA 22203-1977			10. SPONSORING/MONITORING AGENCY REPORT NUMBER F49620-99-1-0061	
11. SUPPLEMENTARY NOTES				
12a. DISTRIBUTION AVAILABILITY STATEMENT APPROVAL FOR PUBLIC RELEASE; DISTRIBUTION UNLIMITED			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) The goal of this research project is to develop novel optical materials and related structures with superior properties for use in nonlinear optics. This work is motivated by the need for high quality materials with large nonlinear coefficients for use in nonlinear and photonics devices and by the realization that new structures developed in recent years provide the possibility for new types of nonlinear optical devices and interactions. Devices that could benefit from these new and improved materials include eye/sensor protection devices, optical switches, and phase conjugating aberration correction devices.				
14. SUBJECT TERMS			15. NUMBER OF PAGES	
20020719 123			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL	

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Final Report

New Materials and Structures for Nonlinear Optics

submitted by

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to

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Grant No. F49620-99-1-0061

Project start date: December 1, 1998

Budget total: \$50,000 per year for three years

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1. Executive Summary of Original Proposal

This document constitutes a request for support for research to be conducted at the University of Rochester as part of a collaboration with research scientists at the Materials and Manufacturing Directorate, Air Force Research Laboratory (AFRL/MLPO) Wright-Patterson AFB. Over the past several years, Prof. Boyd has been developed joint research projects with Robert L. Nelson of this laboratory. Other Air Force scientists interested in this work include Kenneth Hopkins and William R. Woody. The philosophy of this collaboration is that MLPO possesses materials synthesis and processing capabilities that are not typically available at a university laboratory. Moreover, direct collaboration with Air Force personnel will allow Prof. Boyd to develop research themes that are of relevance to the Air Force mission. On the other hand, Professor Boyd can bring to this collaboration extensive experience in the foundations of nonlinear optics and extensive laboratory facilities for performing measurements on nonlinear optical materials developed at WPAFB. At present, we are requesting nominal funding in support of the university portion of this collaboration. Funding at a level of \$50,000 per year is requested to provide partial support of the principal investigator, one graduate or post doctoral assistant, and laboratory equipment and supplies. We are able to conduct research with such a modest budget because of the willingness of MLPO to participate in this collaboration and to provide us with materials for use in this study.

The goal of this research project is to develop novel optical materials and related structures with superior properties for use in nonlinear optics. This work is motivated by the need for high quality materials with large nonlinear coefficients for use in nonlinear and photonics devices and by the realization that new structures developed in recent years

provide the possibility for new types of nonlinear optical devices and interactions. Devices that could benefit from these new and improved materials include eye/sensor protection devices, optical switches, and phase conjugating aberration correction devices.

The central theme of this work entails the development of materials incorporating a composite structure for use in nonlinear optics. Some of the structures that have been successfully used in the design of nonlinear optical materials are illustrated in Fig. 1. The basic idea of this approach is to combine two or more constituent materials in such a manner that the nonlinear optical susceptibility of the composite material exceed those of the constituent materials. Past work performed by ourselves [1,2] and other groups [3-5] has demonstrated that this sort of enhancement of nonlinear optical properties is possible. This enhancement can in many instances be understood as a consequence of local field corrections [6], in which the mixing of various materials to form the composite structure leads to a complicated spatial structure of the local electric field. The resulting inhomogeneity of the local electric field can produce an enhancement of the nonlinear optical response.

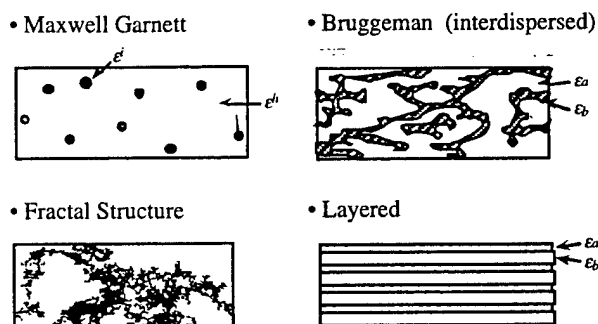


Fig. 1. Composite material structures for use in nonlinear optics.

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1. J. E. Sipe and R. W. Boyd, Phys. Rev. A 46, 1614, 1992.
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3. G.S. Agarwal and S. Gutta Gupta, Phys. Rev. A 38, 5678, 1988.
4. J.W. Haus, R. Inguva, and C.M. Bowden, Phys. Rev. A 40, 5729, 1989.
5. F. Hache, D. Ricard, C. Flytzanis, and U. Kreibig, Appl. Phys. A 47, 347-357 (1988).
6. J. J. Maki, M. S. Malcuit, J. E. Sipe, and R. W. Boyd, Phys. Rev. Lett. 68, 972, 1991.

2. Summary of Results Obtained by this Research Program

The research performed under this contract has led to a greater understanding of the nature of the nonlinear optical response of material systems and led to the theoretical and experimental demonstration of new materials and structures which display large optical nonlinearities. Details are provided in the numerous publications (listed below) resulting from this research grant. Rather than attempting to summarize all of these contributions, in this section we mention two particular successes of this research project.

One of the primary goals of this project was to develop ties between the principal investigator and other scientists working on problems of interest to the US Air Force. During this research project, Dr. Robert Nelson of AFRL was able to put together a

research team comprised of himself and his coworkers at AFRL, Prof. J. Haus at the University of Dayton, Prof. Jay Guo at the University of Michigan, and myself at the University of Rochester. This team was able to obtain seed money from AFOSR. I am presently continuing to collaborate actively with Prof. Guo and less actively with other members of this team. Prof. Guo and I have broad ranging joint interests including the development of organic nonlinear optical materials and the fabrication of polymer photonic devices.

The second example was the demonstration of enhanced electrooptic response of a layered-composite material (see refs 3 and 5 of section 4 below). The experimental setup we used to observe this effect is shown in Fig. 2. In this experiment, we measured the third-order nonlinear response of the composite material and found that it was 3.2 times larger than that of the pure electrooptic material. This result is significant in that it shows that composite materials can be significantly more nonlinear than their constituent ingredients.

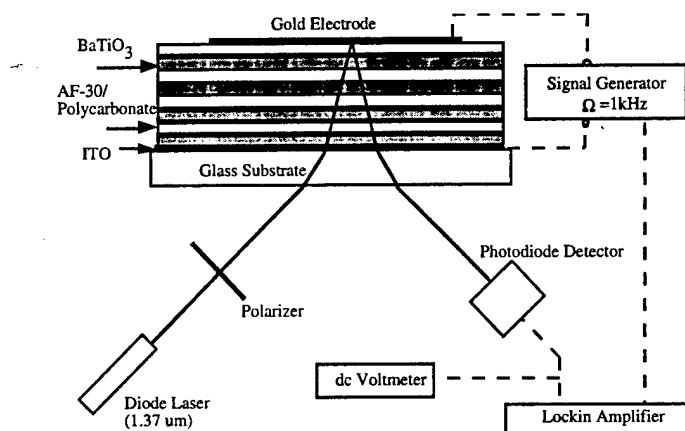


Fig. 2. Experimental setup of Nelson and Boyd used to demonstrate enhanced response of a layered-composite electrooptic material.

3. Prospectives for Further Research

During the course of this research project some ideas came together which are likely to be pursued in the investigator's future research. One of these new ideas is briefly summarized here. It is described more fully in publications 12 and 13 of section 4 of this document.

Nanofabrication of Exotic Optical Materials One future goal is to fabricate materials and structures with optical properties fundamentally unlike those of naturally occurring materials. Some of the exotic characteristics these devices are expected to display include ultra-slow group velocities of propagation, enhanced optical nonlinearities, and large dispersion with a controllable magnitude and sign. Previous related work includes the development of photonic bandgap (PBG) structures and structures consisting of waveguides coupled to arrays of optical resonators. One of the structures to be studied under the present program is shown in part (a) of Fig. 3. It consists of an optical waveguide coupled to a series of optical resonators. The resonators can be of arbitrary

design, although in our experimental work we are concentrating on resonators in the form of a ring waveguide or a whispering gallery mode of a disk. A pulse of light is shown propagating through this structure. Evanescent coupling between the waveguide and resonator injects light into each resonator where it circulates many times (on resonance, $2F/\pi$ times, where F is the finesse of the resonator) before being coupled back into the waveguide. For a densely packed collection of high-finesse resonators, a light wave spends much more time circulating within each resonator than in propagating between resonators. Thus the group velocity of propagation can become very small. Because the time delay acquired in interacting with each resonator depends critically on the detuning of the optical wave from the resonance frequency, this device displays tailorable dispersion with a size that is many order of magnitude larger than that of conventional materials. Also, because of the build-up of intensity within each resonator, the nonlinear response of this structure is greatly enhanced. We have shown [publication 3 of Section 5] that the nonlinear response is enhanced by the square of the resonator finesse in comparison to propagation through a bulk nonlinear material. Under appropriate conditions, these dispersive and nonlinear effects can precisely balance one another, leading to the propagation of optical solitary waves. Some examples of the predicted behavior are shown in Fig. 4.

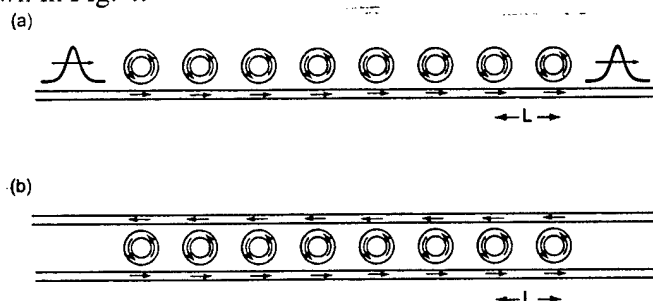


Fig. 3. Structured waveguides to be studied as part of this program and which displays slow light propagation, large controllable dispersion, and large controllable nonlinearity. (a) Single-guide structure and (b) double-guide structure.

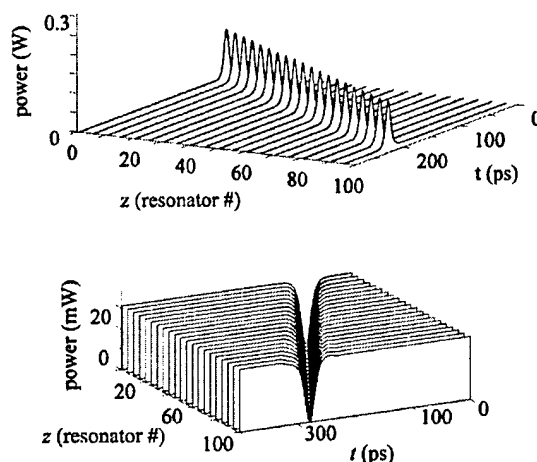


Fig. 4. Because the sign of the group-velocity dispersion for the device shown in Fig. 3(a) can be controlled by varying the optical frequency, both bright (upper panel) and dark (lower panel) optical solitons can be made to propagate through the same physical device.

A related structure is shown in the Fig. 3(b). It consists of upper and lower waveguides coupled to the same sequence of resonators. The optical properties of such a device differ in fundamental ways from those of the single-guide structure. Because forward and backward going waves are coupled in the two-guide structure, it possesses a photonic bandgap, which is not present in the single-guide structure. Both structure are expected to possess useful photonic properties, which however differ in their detailed behavior. Within the present research program we will study the fundamental optical properties of structures of this sort, although we expect them to have important implications for applications such as soliton propagation, controllable optical delay lines, and photonic switching.

Fabrication Considerations of Structured Waveguides. We have become quite familiar with the fabrication of photonic devices as part of a separate funded program aimed at the fabrication of photonic sensors for the detection of biological pathogens. Waveguiding GaAs/AlGaAs structures are grown in Rochester using the MBE facility of Prof. Wicks. The horizontal patterning is performed using state-of-the art nanofabrication procedures and is taking place at the Cornell Nanofabrication Facility (CNF), a user facility funded largely by US government support. Fabrication is proceeding quite well. Fig. 5 shows a device after being transferred into the GaAs/AlGaAs waveguiding structure. The particular device that is illustrated consists of several resonators coupled to the same waveguide. We are presently in the process of characterizing the optical properties of the devices fabricated to date.

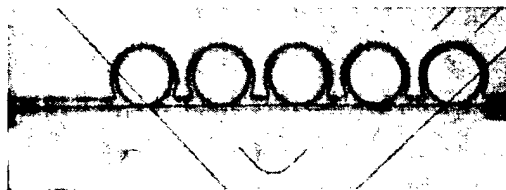


Fig.5. Optical photograph of a completed ring-coupled waveguide device.

4. Publications on Nonlinear Optical Materials Resulting from this Research Grant

1. Photorefraction in Lead-Tin Fluorophosphate Glass, S. Radic, R. J. Essiambre, R. W. Boyd, P. A. Tick, and N. F. Borrelli, *Opt. Lett.* 23, 1730, 1998.
2. Order-of-Magnitude Estimates of the Nonlinear Optical Susceptibility, R. W. Boyd, *Journal of Modern Optics*, 46, 367, 1999
3. Enhanced Electrooptic Response of Layered Composite Materials, R. L. Nelson and R. W. Boyd, *Applied Physics Letters*, 74, 2417, 1999.
4. Enhanced Third-Order Nonlinear Optical Response of Photonic Bandgap Materials, R. L. Nelson and Robert W. Boyd, *Journal of Modern Optics*, 46, 1061, 1999.
5. Accessing the Optical Nonlinearity of Metals with Metal-Dielectric Photonic Bandgap Structures, R. S. Bennink, Y.-K. Yoon, R. W. Boyd, and J. E. Sipe, *Opt. Lett.* 24, 1416, 1999.
6. Enhanced Electrooptic Response of Layered Composite Materials, R. L. Nelson and R. W. Boyd, Highlighted in the Optics in 1999 issue of *Optics and Photonics News*, December 1999.

7. Z-Scan Measurement of the Nonlinear Absorption of a Thin Gold Film, D. D. Smith, Y. K. Yoon, R. W. Boyd, J. K. Campbell, L. A. Baker, R. M. Crooks, and M. George, *J. Appl. Phys.*, 86, 6200, 1999.
8. Intrinsic Optical Bistability in a Thin Layer of Nonlinear Optical Material by means of Local Field Effects, Y. K. Yoon, R. S. Bennink, R. W. Boyd, and J. E. Sipe, *Optics Communications*, 179, 577, 2000.
9. Nonlinear Optical Materials, R. W. Boyd and G. L. Fischer, an article to be published in the *Encyclopedia of Materials: Science and Technology*, Pergamon Press, Oxford 2000.
10. Nanocomposite Materials for Nonlinear Optics, J. E. Sipe and R. W. Boyd, an article to be published in *Handbook of Advanced Electronic and Photonic Materials*, edited by H. S. Nalwa, Academic Press, 2000.
11. Measurement of the thermal contribution to the nonlinear refractive index of air at 1064 nm, S. J. Bentley, R. W. Boyd, W. E. Butler, and A. C. Melissinos, *Opt. Lett.* 25, 1192, 2000.
12. Slow Light, Induced Dispersion, Enhanced Nonlinearity, and Optical Solitons in a Nanostructured Waveguide, J. E. Heebner, R. W. Boyd, and Q-H. Park, *Phys. Rev. E*, *****, 2002.
13. SCISSOR Solitons and Other Novel Propagation Effects in Fully Transmissive Structured Waveguides, J. E. Heebner, R. W. Boyd, and Q-H. Park, *J. Opt. Soc. Am., B*. *****, 2002.

5. Investigator's Other Publications During This Funding Period

1. Preventing Laser Beam Filamentation Through Use of the Squeezed Vacuum, R. W. Boyd and G. S. Agarwal, *Phys. Rev. A*, 59, R2587, 1999.
2. Elimination of the Bandgap of a Resonant Optical Material by Electromagnetically Induced Transparency, G. S. Agarwal and R. W. Boyd, *Phys. Rev. A*, 60, R2681, 1999.
3. Enhanced All-Optical Switching by use of a Nonlinear Fiber Ring Resonator, J. E. Heebner and R. W. Boyd, *Opt. Lett.* 24, 847, 1999.
4. Conversion of Unpolarized Light to Polarized Light with Greater Than 50% Efficiency by Photorefractive Two-Beam Coupling, J. E. Heebner, R. S. Bennink, R. W. Boyd, and R. A. Fisher, *Optics Letters* 25, 257, 2000.
5. Photorefractive Optical Recycling for Contrast Enhancement, J. E. Heebner and R. W. Boyd, *Optics Communications*, 182, 243-247, 2000.
6. Efficient Infrared Imaging Upconversion via Quantum Coherence, R. W. Boyd and M. O. Scully, *Appl. Phys. Lett.* 77, 3559, 2000.
7. The Impact of Charles H. Townes on Nonlinear Optics, R. W. Boyd, *IEEE LEOS Millenium Issue of the Journal of Selected Topics in Quantum Electronics*, 6, 881-884, 2000.
8. Enhanced self-action effects by electromagnetically induced transparency in the two-level atom, R. S. Bennink, R. W. Boyd, C.R. Stroud, Jr., and V. Wong, *Phys. Rev. A* 63, 033804 (2001).
9. Modification of Self-Induced Transparency by a Coherent Control Field Q-Han Park and R. W. Boyd, *Phys. Rev. Lett.* 86, 2774 (2001).
10. Comment on "Quantum Interferometric Optical Lithography: Exploiting Entanglement to Beat the Diffraction Limit," G. S. Agarwal, R. W. Boyd, E. M. Nagasako, S. J. Bentley, *Phys. Rev. Lett.*, 86, 1389, 2001.

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13. Spatial patterns induced in a laser beam by thermal nonlinearities, S. J. Bentley, R. W. Boyd, W. E. Butler, and A. C. Melissinos, *Opt. Lett.* 26, 1084-6, 2001.
14. Absorptionless Self-Phase Modulation in a Dark-State Electromagnetically Induced Transparency System, V. Wong, R. W. Boyd, C. R. Stroud, Jr., R. S. Bennink, D. L. Aronstein, and Q.H. Park, *Phys. Rev. A* 65, 013810, 2001.
15. Parametric downconversion vs. optical parametric amplification: a comparison of their quantum statistics, E. M. Nagasako, S. J. Bentley and R. W. Boyd, and G. S. Agarwal, *J. Mod. Optics*, 49, 529 2002.
16. Honeycomb Pattern Formation by Laser-Beam Filamentation in Atomic Sodium Vapor, R. S. Bennink, V. Wong, A. M. Marino, D. L. Aronstein, R. W. Boyd, C. R. Stroud, Jr., S. Lukishova, and D. J. Gauthier, *Phys. Rev. Lett.* 88, 113901, 2002.
17. "Slow" and "Fast" Light, R. W. Boyd and D. J. Gauthier, a book chapter to appear in *Progress in Optics*, 2002.

6. Special Mention

During the period of this award the following events occurred

- (a) Prof. Boyd was elevated to the position of Fellow in the American Physical Society
- (b) Prof. Boyd was promoted to a chaired professorship at the University of Rochester
- (c) One of Prof. Boyd's papers was highlighted on the cover of *Physical Review Letters*
- (d) Prof. Boyd served as the Chair of the Division of Laser Science of the American Physical Society